Vortex Pinning Centers in High-Temperature Superconducting Films

Subjects: Materials Science, Coatings & Films
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To better pin the vortex at external magnetic fields, the HTS films must contain APCs with desired morphology, dimension, orientation, and concentration. Nanoscale APCs with lateral dimension approaching 2ξ (coherence length) on the order of a few nanometers in HTSs must be generated to suppress the dissipation of vortex motion. This has prompted extensive efforts and exciting results have been obtained in generating nanoscale APCs in HTS films. The research progress of different types and dimensions APCs in detail is introduced and the impact on superconducting performance is summarized.

Keywords: high-temperature superconducting films ; vortex pinning ; artificial pinning center ; nanoparticle ; nanocolumn

1. Introduction

Superconductors are a class of materials with unique physical properties and high application value. Within critical parameters, superconductors have two major properties, namely: the zero-resistance effect and the Meissner effect. T_c (critical transition temperature), B_c (critical magnetic field), and J_c (critical current density) are the main critical parameters of superconductors. Currently, superconductors are classified into Type-I and Type-II superconductors. For the Type-I superconductor, there is only one critical magnetic field B_c. However, for the Type-II superconductor, there are two critical magnetic fields, the lower B_{c1} and upper B_{c2}. When $B_{c1} > B$, the superconductor remains in the Meissner state, completely expelling the magnetic flux from its interior. For $B_{c2} > B > B_{c1}$, the magnetic flux starts penetrating the sample in the form of discrete bundles termed “flux lines” and the sample goes into the mixed state (or vortex state). When $B > B_{c2}$, the superconductor comes into the normal state.

Usually, Type-II superconductors that exhibit superconducting states at ~30 K and above are called high-temperature superconductors (HTSs). They are first discovered in 1986 by J. G. Bednorz and K. A. Müller in the Ba-La-Cu-O system [1]. $YBa_2Cu_3O_{7-\delta}$ (YBCO) is the first HTS to be found with T_c above the liquid nitrogen temperature (LN_2, 77 K) [2], and its discovery triggers a research boom in the field of superconductivity. Early research on HTS focused on exploring superconductors with higher T_c, and scientists subsequently discovered several systems such as $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (BSCCO) [3], $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ (TlBCCO) [4], $HgBa_2Ca_2Cu_3O_{8+\delta}$ (HgBCCO) [5], all with T_c~77 K and above. Since HTSs allow the use of LN_2 as a cooling source, which is readily available and relatively inexpensive, they have a significant cost advantage over low-temperature superconductors (LTS) for future large-scale practical applications [6][7]. Mahesh Paidpilli et al. summarized in detail the various applications of HTSs in the high magnetic field in the United States at present in their review [8]. D. Uglietti summarized relevant research and developments in commercial HTS materials applied in large solenoids, accelerator dipoles, and high-field tokamaks [9]. In addition, other prominent applications of HTS including single-photon detectors based on superconducting nanowires (SNSPDs) [10], superconducting quantum interference devices (SQUIDs) [11][12][13] et al. have also been reported.

For HTS films, commonly used substrates include MgO, $SrTiO_3$, $LaAlO_3$, $LaSrAlO_4$, YSZ (Yttria-Stabilized Zirconia), sapphire (with Ag, CeO_2, and MgO buffer layers), and so on [14][15]. Due to the structural complexity, the phase composition of HTS films can vary depending on the deposition methods and parameters (e.g., substrate types, temperature, vacuum quality, accelerating voltage, etc.), resulting in different calcined phases. In addition, individual elements may be present in the form of metal oxides or compounds, and the generation of these additional phases increases the difficulty of preparing HTS films [16][17][18][19][20][21][22].

2. Artificial Pinning Centers (APCs)

To better pin the vortex at external magnetic fields, the HTS films must contain APCs with desired morphology, dimension, orientation, and concentration. Nanoscale APCs with lateral dimension approaching 2ξ (coherence length) on the order of a few nanometers in HTSs must be generated to suppress the dissipation of vortex motion. This has prompted extensive efforts in the past few decades or so and exciting results have been obtained in generating nanoscale APCs in HTS films. In this section, researchers introduced the research progress of different types and dimensions APCs in detail, and summarized the impact on superconducting performance.

2.1. Zero-Dimensional APCs (0D APCs)

The effect of ionic radii on the T_c of *RE*BCO has been documented in previous work [23]. It is well known that varied rare-earths have different ionic radii. The phenomenon that T_c varies linearly with ionic radius of *RE* ions has been detected and was attributed to strain-induced charge redistribution between the CuO_2 planes and the charge reservoir (CuO-chains). Several rare-earth elements, including Sm, Eu, and Nd, have been doped in place of Y with various molar cationic ratios to enhance the vortex-pinning capabilities of YBCO films [24]. The Y atom in Y-Ba-Cu-O has been totally replaced in certain studies [24][25][26][27] by another rare-earth atom or a mixture of two or more rare-earth atoms, which has improved vortex pinning. Several combinations, including ($Gd_{0.8}Er_{0.2}$) [26] and ($Nd_{1/3}Gd_{1/3}Eu_{1/3}$) [27], were published to determine whether the strain caused by lattice mismatch increased when mixtures of rare-earth elements were used instead of a single rare-earth element. Except for the situation when defects were random and unrelated, the enhancement was not notable in any circumstances. There have been attempts to substitute Tb, Ce, Pr, Nd, La, Co, Dy, and Eu at the Y site of YBCO and the *RE* site of *RE*BCO films [28][29][30][31][32]. The increased density of these substituent nanoprecipitates in doped *RE*BCO films compared to pristine *RE*BCO film led to elevated J_c and F_p values across a wide range of applied magnetic fields, which in turn led to stress field due to lattice mismatch between the phases in the resulting *RE*BCO films.

2.2. One-Dimensional APCs (1D APCs)

The idea of strain engineering has been applied to generate and control the morphology and dimension of APCs embedded in HTS films. According to the elastic strain energy model, the appropriate level of interfacial strain can act as a driving force for the self-assembly of 1D vortex pinning, controlling the morphology [33][34], dimensionality [35][36], orientation [37], and concentration [38]. Numerous studies have shown that 1D columnar APCs grown along the *c*-axis of $REBa_2Cu_3O_{7-\delta}$ films exhibit strong vortex pinning ability, resulting in high J_c when the applied magnetic field is along the *c*-axis direction [33][38][39][40].

MacManus-Driscoll et al. [41] first reported the introduction of $BaZrO_3$ secondary phase into YBCO films using the PLD (Pulsed Laser Deposition) technique to enhance the performance. It was found that $BaZrO_3$ nanoparticles and nanocolumns produced significant *c*-axis orientation-related enhancement of J_c despite its random distribution in the YBCO matrix. Following the work of MacManus-Driscoll et al., 1D $BaZrO_3$ APCs have been intensively investigated. In the subsequent report by Yamada et al. [42], the addition of YSZ (yttrium oxide stabilized zirconium oxide) to YBCO targets resulted in the formation of columnar $BaZrO_3$ nanostructures in YBCO films and would leave a YBCO film matrix containing Ba defects. Self-assembly of vertical arrays of $BaZrO_3$ phases is observed in this composite film. The vertical alignment of these self-assembled $BaZrO_3$ columnar phases was hypothesized to be due to the preferential nucleation of impurity islands in the strain field above the impurity particles [43]. Physical property measurements showed that these self-assembled vertical $BaZrO_3$ phase arrays resulted in strong pinning of vortices, especially when the applied magnetic field was along the *c*-axis direction. Goyal et al. [44] also reported enhanced pinning of $BaZrO_3$/YBCO nanocomposite films along the *c*-axis direction. The $BaZrO_3$/YBCO interface is strongly strained due to the high lattice mismatch of 7.7% between $BaZrO_3$ and YBCO, which leads to the formation of a high defect density semi-coherent $BaZrO_3$/YBCO heterointerface [45][46]. This defect is considered the source of the high pinning efficiency achieved at the 1D $BaZrO_3$ magnetic flux pinning centers.

The search for new vortex pinning materials with smaller lattice mismatches with HTSs is the most effective and likely solution to improve superconductivity. In addition to $BaZrO_3$, 1D-nanostructured materials such as $BaSnO_3$ [47][48][49][50][51][52][53][54][55], $BaTiO_3$ [56], $BaHfO_3$ [47][57][58][59], $YBa_2(Nb/Ta)O_6$ [60][61][62][63] have also been successfully introduced into YBCO films using the PLD technique. These 1D APCs provide different degrees of vortex immobilization [47][52][57][58][59][61][62][64]. In all cases, the enhancement of J_c is more pronounced when the applied magnetic field is higher. Mele P et al. [55] reported a record $F_{p,\ max}$ value of 28.3 GN/m^3 for $BaSnO_3$/YBCO nanocomposite films, reflecting the excellent J_c performance at that time. In addition, the double-perovskite material, YBa_2NbO_6 (YBNO), was also investigated as 1D APCs and introduced into the superconducting matrix [65]. In another study, Jha A. K. et al. [66] applied surface-modified target method to introduce YBa_2NbO_6 columns into YBCO films by controlling the rotational speed of the target to control the concentration of YBa_2NbO_6. YBa_2NbO_6 nanocolumns were observed to effectively enhance the J_c performance of YBCO films. Furthermore, RE_3TaO_7 and $REBa_2TaO_6$ were also proved to significantly enhance the J_c performance of

*RE*BCO films [67][68], and the results indicated that lattice mismatch is a suitable condition to produce high pinning ability in the range of 5~12% [68].

Recently, $BaHfO_3$(BHO) has sparked much interest among researchers as a very promising secondary phase APC, whose nano-inclusions in the form of columnar or spherical structures within the *RE*BCO matrix significantly improve the J_c values of *RE*BCO films deposited on single crystals and metal strips [69][70][71][72][73][74][75][76]. Tobita et al. [69] firstly reported that the BHO-doped $GdBa_2Cu_3O_y$(GdBCO) film was deposited by PLD on the IBAD-MgO substrate. The most interesting feature of BHO nanocolumns addition was reported as J_c is undepressed by increasing thickness of the film. By using the LTG (Low-Temperature Growth) technique in PLD, $BaHfO_3$/SmBCO films exhibit very high $F_{p,\ max}$ (~28 GN/m^3) at 77 K when H is parallel to the *c*-axis [73]. Even on metal tapes, the $BaHfO_3$/GdBCO nanocomposite films exhibit a large $F_{p,\ max}$ (~23.5 GN/m^3) and a high irreversibility field ($\mu_0H$irr = 15.8 T) when H is parallel to the *c*-axis at 77 K [74]. In addition, $BaHfO_3$ nanoparticles were also introduced into YBCO [75] and GdBCO [76] films using the CSD method, which improved the J_c of the nanocomposite films.

2.3. Two-Dimensional APCs (2D APCs)

The deposition of multilayer or quasi-multilayer film structures has also been used in HTS films to improve vortex pinning capabilities. For example, YBCO multilayer films have been prepared using the PLD technique (intermediate layers include: Ag [77], Pd [78], Y_2O_3 [79][80][81], $BaZrO_3$ [82][83], $SrRuO_3$ [84], $SrTiO_3$ [85], $LaCaMnO_3$ [86], YSZ [87], Y-211 [88], $PrBa_2Cu_3O_x$ [89] and transition metals Ir [90], Ti, Zr, Hf [91]). The formation of $BaMO_3$ (M = Ti, Zr, Hf, Ir) phases can be observed after the addition of transition metal elements to YBCO films. Not only the J_c enhancement based on YBCO multilayers was observed from the physical property test results, but also the irreversibility lines moved to higher *H*-*T* regions [82].

2.4. Three-Dimensional APCs (3D APCs)

To obtain better performance, the ideal materials need to be carefully selected when introducing the secondary phase to the HTS matrix. In terms of pinning efficiency, the spherical secondary phase needs to maintain the proper size and shape, and is required to be uniformly distributed among the superconducting matrix, which is necessary. Therefore, it is not an easy task to find a secondary phase material that will persist in ideal presence and distribution during superconductor synthesis as APCs. At present, many compounds have been applied to investigate the possibility of becoming effective APCs.

2.5. Hybrid 1D + 3D APCs

The 1D columnar APCs perform very well in enhancing J_c, but one of the shortcomings is that the performance of J_c degrades more with the change of direction of applied external magnetic fields. In addition, at higher temperatures, due to thermal excitation, the vortex tends to form a double kink structure, and even if they contain crystal defects in the *c*-axis direction, the unpinned vortices can still move due to the Lorentz force, resulting in degraded performance. To solve this problem, combinations of APCs with different dimensions have been developed. It has been shown that the simultaneous formation of 1D and 3D APCs can effectively compensate for the lack of performance of 1D columnar APCs only, adapting to applied magnetic fields with different applied directions [92].

Mele et al. [93] reported the combined application of two different types of pinning centers to successfully introduce both $BaZrO_3$ columns and Y_2O_3 nanoparticles into YBCO films using the PLD technique. Although J_c increased only slightly in the intermediate angular region, the significant decrease of J_c with angle in *c*-axis direction was significantly improved compared to YBCO films with only $BaZrO_3$ nanocolumns added. Similar results were obtained in a related study by Ding F. Z et al. [94]. Subsequently, combinations of columns with nanoparticles of different materials were also reported to enhance the J_c performance of YBCO films, sufficiently reducing the anisotropy of J_c [49][61][95][96]. For example, $BaSnO_3$ columns and Y_2O_3 nanoparticles were tried in combination, which significantly enhanced J_c and reduced the anisotropy of J_c [48][96]. TEM studies of YBCO + 3%$BaSnO_3$ and YBCO + 3%$BaSnO_3$ + Y_2O_3 nanocomposite films showed that only columnar nanostructures were formed in YBCO + $BaSnO_3$ films, while YBCO + $BaSnO_3$ + Y_2O_3 thin films formed both columnar and spherical nanostructures.

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